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Risk assessment for human health in a seleniferous area, Shuang'an, China

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Abstract Shuang'an in Ziyang is a typical high-selenium (Se) area in China, where human selenium (Se) poisoning was reported 30 years ago. To assess the risk of Se poisoning in the area, the Se content in agricultural soil, plant, and water in Naore, Shuang Hekou, and Lin Benhe villages of Ziyang was systematically investigated. The probable daily intake (PDI) was calculated on the basis of Se contents in food, water, and dietary habits to evaluate the risk of selenosis. Se content in hair from local habitants (32 men and 34 women) was determined to further verify the risk. Results showed that Se content in the soil ranged from 0.21 to 36.07 mg/kg, with a geometric means of 3.02 ± 5.16 mg/kg, respectively. Approximately 60% of soil reached the Se toxicity threshold (>3 mg/kg). The Se content in plants ranged from 0.02 to 17 mg/kg, with an average of 0.76 ± 2.51 mg/kg. Approximately 40% of the plant reached the toxicity standard (1 mg/kg). Se content in soil and plant of Naore village was significantly higher than that from two adjacent villages. The Se content in vegetables (on fresh weight basis) was higher than that in cereals. Se contents were 3.73 ± 9.08 and 1.32 ± 3.50 mg/kg in eggplant and pepper, respectively. The Se content in drinking water was 7.85 ± 6.04 µg/L, lower than the upper tolerable limit (40 µg/L) set by WHO. The Se content in stream water (18.5 µg/L) was significantly higher than that in drinking water because of soil erosion. The calculated PDI of habitants in Naore village (1801 µg/day) was significantly higher than that in Lin Benhe (666 µg/day) and Shuang Hekou (686 µg/day), all of which was higher than the tolerable upper Se intake (400 µg/day) set by WHO. The calculated blood Se content was relatively high, especially for habitants of Naore village (2783–2824 µg/L). Moreover, the Se content in the hair of local habitants reached as high as 12.72 ± 13.81 mg/kg, and 78.79% exceeded hair Se toxicity threshold (>3 mg/kg), which further verified the potential selenosis risk. Our study provides significant implications of the potential Se intoxication of local residents. Therefore, governments and other institutions should implement various measures to reduce the daily Se intake and to mitigate the associated health risks.

Keywords Selenium · Ziyang · Soil and water · Food · Daily Se intake · Risk assessment

Introduction

Selenium (Se) is an essential element for human health, and the most of natural intake is usually from diet (Chilimba et al. 2011). In recent years, this element has been applied to treat and prevent severe pathologies, such as cancer, cardiovascular disorders, thyroid adenoma, and osteodystrophia, among residents living in Se-deficient areas (Burk 2002; Thiry et al. 2012). Se can also be toxic at high concentration, and its toxicity is characterized as acute and chronic (Yang et al. 2013). Vomiting and diarrhea are often present as acute toxicity, usually accompanied by rapid heartbeat (ATSDR 2003). Hair and nail lose are the prime symptoms of chronic toxicity (Yang et al. 1983), which may also be associated with skin lesions and disorders of the nervous system (Nuttall 2006).

Severe selenosis can lead to weakness, paralysis, and even death (Fordyce 2007). Thus, local habitants may suffer from severe Se poisoning over time when high Se contents exist in the environment (Hira et al. 2004).

Toxic Se levels in food chain accumulate via soil and water that cause serious health problems (Fordyce 2007; Yang et al. 2013); this phenomenon was found in some countries, such as China, USA, India, Canada, Mexico, New Zealand, and Australia (Yang et al. 1989; Aro et al. 1995; Navarro-Alarcon and Cabrera-Vique 2008; Combs 2001; Fordyce 2013). For example, high Se content in water and soil results in local inhabitants facing a high risk of Se poisoning via having Se-rich foods in Enshi seleniferous area of China as reported by Qin et al. (2013). In addition, Ziyang seleniferous region is another notable Se-excessive or seleniferous regions in China. Se toxicity symptoms occurring in Enshi were also observed in Ziyang in 1984, and the most severely affected area was Naore village of Shuang'an County (Mei 1985; Fang and Wu 2004). The reported Se content in soil of Naore village was 15.74 mg/kg (Mei 1985), 16.9 mg/kg (Fang et al. 2003), and 26 mg/kg (Zhao et al. 1993), respectively. Mei (1985) attributed the high soil Se content to a parent compound, and Luo et al. (2003) also demonstrated that high Se content in soil is mainly due to Se-rich pyritic carbonaceous slate and volcanic substances under a thin soil layer.

Se content and availability in soil determine Se levels in plants and water, which determine the Se status in the human body. Besides, Se levels in various plants differ because of diverse geological conditions, growth conditions, absorptive capacities, growth periods, and protein contents (Levander 1987, 1982; Temmerman et al. 2014); Finglas 2000; Yu et al. 2015). Therefore, the dietary Se intake of humans worldwide considerably varies because of different Se contents in food and various dietary structures (Mcnaughton and Marks 2002). The recommended dietary allowance (RDA) of Se is 55 μg/day by WHO (1986), which are 70 and 50 μg/day for males and females, respectively, by the US Food and Nutrition Board (1980). No observed adverse effect levels (NOAELs) set by WHO (1986) are 240 μg/day (4 μg/kg bw/day), and upper tolerable level set by the Institute of Medicine (IOM 2000) is 400 μg/day. Accordingly, the Se status in humans should be evaluated within the narrow range between moderate and toxic levels. Probable daily intake (PDI) widely ranged from <10 μg/day for people living in Se-deficient areas to 4990 μg/day for habitants living in selenosis-dense areas in Enshi (Yang et al. 1983). Dhillon and Dhillon (1991) and Hira et al. (2004) examined environmental Se levels and health conditions of individuals inhabiting seleniferous areas in India and found that long-term consumption of natural Se-rich crops and forage leads to high Se intake in humans (PDI of males and females = 632 ± 31.2 and 475 ± 52.8 μg/day, respectively).

However, signs and symptoms of Se poisoning in Ziyang had been reported 30 years ago; no systematical studies have documented the Se levels in food and water, and levels of Se exposure of local populations. In our study, the total Se contents in soil, food, water, and human hair were determined in this area: (1) to investigate the Se status in soil, crops, and water from a Se-contaminated village and two other nearby villages; (2) to estimate PDI and to assess the potential risk of Se poisoning of local dwellers; and (3) to further verify Se status of inhabitants by levels in human hair. In addition, this study was conducted with an aim to provide a reference for the improvement of the health conditions of individuals living in Se-toxic areas.

Materials and methods

Sample collection

Two sampling campaigns were carried out. (1) In 2009, 26 soil samples, 16 corn samples, 7 rape seeds, 6 rice samples, and 18 water samples were collected from Naore village; 21 hair samples (male = 13; female = 8) were also collected. (2) In 2012, 138 sampling sites for soil samples were set in three villages (Naore (NR), Lin Benhe (LBH), Shuang Hekou (SHK)) in Shuang'an countryside in Ziyang; corresponding plant samples of 103 soil sampling sites and other 151 plant samples were also collected in these three villages. Stream water ($n = 29$), drinking water ($n = 10$), lotus pond water ($n = 4$), and hair of residents ($n = 45$, male = 19; female = 26) were also collected to estimate the daily dietary intake and for risk assessment; approximately 2.0 g of hair samples (about 1–3-cm length) was collected from the nape to the scalp (or occipital) region during the interview and investigation on habitants. For soil samples, five samples were obtained randomly from a 20 m × 20 m unit, and the five topsoil (0–20 cm) samples were mixed to prepare a composite sample for each site. Longitude and latitude, as well as the altitude of each sampling site, were recorded by GPS (Fig. 1). The edible parts of fruits and vegetables were collected for analysis. Water samples were obtained from rivers, irrigation channels, and residential water wells.

Sample preparation and analysis

Soil samples were air-dried, ground, sieved to <0.15 mm, and stored in plastic bags before analysis was conducted. Cereal samples were washed with tap water to remove soil and dust, and then washed with deionized water to scour the samples completely. The fresh weight of plant samples was measured; after that, the enzymes of the cereal samples were deactivated at 90 °C for 30 min and oven-dried at 60 °C to a constant weight. After the samples were ground, the cereal samples

Fig. 1 Location of the study area and distribution of the sampling sites
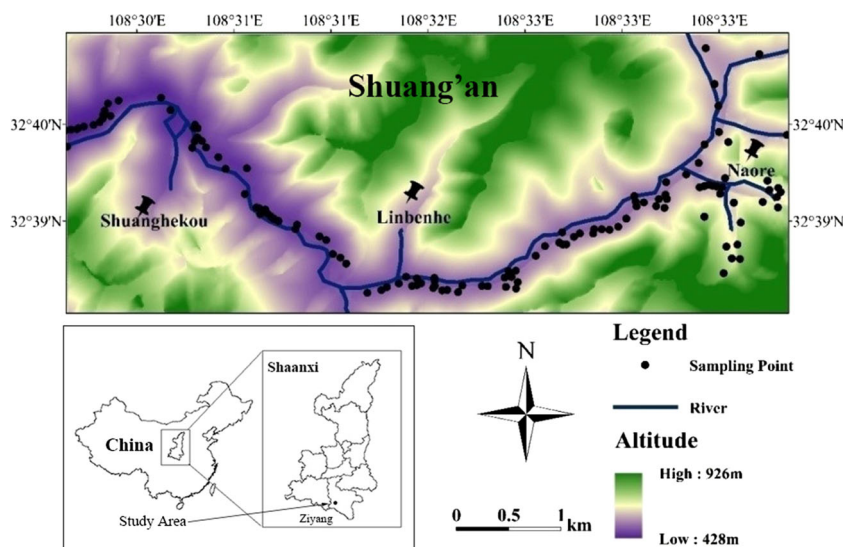
were stored in polyethylene plastic bags at 25 °C for analysis. Fruits and vegetables were washed with deionized water, air-dried, pulverized with a beater, and stored. HNO$_3$ was used to acidify the water samples that were then preserved in a refrigerator. Hair samples were washed with detergent and ethanol and subsequently stored in polyethylene plastic bags.

Total Se levels in soil, plant, and hair samples were determined in accordance with previously described methods (Gao et al. 2011; Wang et al. 2012a), with some modifications. Different HNO$_3$–HClO$_4$ proportions were used to digest soil (3:2, v/v), plant samples (dried), and hair samples (4:1, v/v). Food samples and plant samples were digested the same way, while water samples used a mixture of concentrated HNO$_3$ and 30% H$_2$O$_2$ under conditions similar to those of soil samples. Soil and plant samples were treated and analyzed in accordance with a previously described method (Yu et al. 2015).

The Se content was determined through hydride generation atomic fluorescence spectrometry (AFS-930) in accordance with the standard method (GB/T 5009.93-2003; Ministry of Health of the People's Republic of China 2010); the method detection limit was 0.1 μg/L.

Quality control and statistical analysis

Quality control was assessed using Chinese national certified reference materials, duplicates, and matrix spike quality control samples; an acidified Millipore water sample was also taken as laboratory control. GBW07404 (limestone soil, Se = 0.64 ± 0.18 mg/kg), with a Se content of 0.65 ± 0.06 mg/kg (n = 3), was used as the standard soil reference material, while GBW10014 (cabbage leaves Se = 0.21 ± 0.01 mg/kg) with a Se content of 0.22 ± 0.02 mg/kg (n = 3) was used as standard plant reference material. The relative percentage difference of sample duplicates was <10%.

Calculation

Bioaccumulation factor

The bioaccumulation factor (BAF) was an indicator to evaluate the transfer of Se from soil to plants. The BAF can be calculated as the following:

$$BAF = C_p/C_s \tag{1}$$

Estimation of daily dietary Se intake and hazard quotient

The daily dietary intake was calculated using the following equation:

$$PDI = \sum \left(C_{Se}^i \times IR \right) \tag{2}$$

where PDI is the probable daily intake of Se, C_{Se}^i is Se content in food, and IR is the amount of each foodstuff consumed per capita per day, collected from the Shaanxi Statistical Yearbook 2015; in this book, the consumption data for each type of foods by local habitants were shown specifically.

Hazard quotient (HQ), which evaluates the non-carcinogenic risk of Se for dwellers in the study area, was calculated by dividing the average daily Se intake (ADSel) by the reference dose (RfD). HQ values equal to or less than 1 meant that the risk of Se poisoning can be neglected; however, an HQ value higher than 1 meant that Se exposure estimates exceeded the limit bringing elevated health risks.

$$Hazard\ quotient\ (HQ) = \frac{ADSel\ (\mu g/kg\ bw/day)}{RfD\ (\mu g/kg\ bw/day)} \tag{3}$$

ADSel was calculated by assuming average body weight (bw) to be 70 kg/adult, while the reference dose was calculated as 3.4 μg/kg/day based on the NOAEL (240 μg/kg/day) (Emmanuelle et al. 2012).

Estimation of Se content in blood

According to previous research, Se levels in the blood can be predicted through two equations: Eq. (4), which is based on the relationship between human Se intake (Baoyan and Xuelin 2000) and blood Se content, and a pharmacokinetic model Eq. (5), which is calculated using daily Se intake (Chien et al. 2003; Mahapatra et al. 2001). However, the estimated blood Se content can be confirmatory only when calculated values of Eqs. (4) and (5) are similar to each other (Qin et al. 2013).

$$\log BSe = 0.767 \times \log PDI - 2.046 \tag{4}$$

$$\tau = (C \times V)/(PDI \times F) \tag{5}$$

where BSe is the Se content in blood (μg/L), PDI is the daily Se intake (μg/day), τ is the turnover time of Se (17 days), C is the steady-state concentration of Se in blood (μg/L), V is the volume of blood in the body (5.5 L), and F is the uptake fraction into blood through ingestion (50%).

Statistical analysis

The normality of Se concentration in soil, water, plant, and human hair was assessed using the Kolmogorov–Smirnov test. The logarithmetic transformations of log-normally distributed data were applied for further statistical analysis. The variance of the transformed data was performed by means of a one-way analysis of variance (ANOVA) using the Tukey HSD multi-comparisons (SPSS, version 23). Variance was considered significant for $P < 0.05$.

Results

Se content in the environment of Shuang'an, Ziyang

Se in soil

Se content in soil ranged from 0.21 to 36.06 mg/kg, with geometric mean of 3.02 ± 5.16 mg/kg. Soil Se content from Naore village, where the selenosis symptom was reported in 1980s, was the highest ($n = 64$, geometric mean = 3.79 ± 8.56 mg/kg), which was significantly higher than that in SHK ($n = 46$, geometric mean = 2.60 ± 1.89 mg/kg) and LBH villages ($n = 49$, geometric mean = 1.60 ± 1.60 mg/kg, $P < 0.01$, Fig. 2). Naore

village had the most uneven Se distribution in soil with the variation coefficient reached 120%.

Based on the abundance and deficiency indexes proposed by Tan (1989), 54.09% of the soil samples were Se toxic (>3 mg/kg), and the proportions of Se-enriched (0.40–3.0 mg/kg) and moderate soil samples (0.18–0.40 mg/kg) were 14.47 and 23.90%, respectively. The proportions of toxic soil (3 mg/kg) from NR, SHK, and LBH villages were 71.88, 56.52, and 28.57%, respectively. Approximately 97, 93, and 86% of the soil samples from the NR, SHK, and LBH villages, respectively, exceeded the adequate Se standard (0.40 mg/kg).

Se in plants

Se content in different kinds of plants ranged from 0.02 to 17 mg/kg with a geometric mean of 0.76 ± 2.51 mg/kg (Table 2). The highest Se content (0.81 ± 3.12 mg/kg) was observed in corn, but no significant difference was found among cereal plants. The highest Se content was observed in eggplant, garlic, and pepper among vegetables (Table 1). Se content of 91 samples (36%) exceeded the Se toxicity standard (>1 mg/kg) proposed by Tan (1989). Se content in plant collected from NR ($n = 81$; geometric mean 1.25 ± 4.04 mg/kg) was significantly higher than that from LBH ($n = 77$; geometric mean 0.47 ± 1.77 mg/kg) and SHK ($n = 93$; geometric mean 0.48 ± 2.75 mg/kg) ($P < 0.05$). The proportions of plant with toxic Se level from NR, LBH, and SHK villages were 55.56, 29.87, and 23.66%, respectively.

Se in water

Se contents from streams, wells, ponds, and rice fields of three villages ranged from 1.86 to 36.82 μg/L with a geometric mean of 12.35 ± 9.45 μg/L. Se content was higher in stream water ($n = 35$, geometric mean 15.54 ± 10.46 μg/L) than in pond water ($n = 4$, geometric mean 15.03 ± 3.5 μg/L), rice field water ($n = 5$, geometric mean 9.88 ± 0.39 μg/L), and potable water ($n = 17$, 7.85 ± 6.04 μg/L). Se content in potable water of NR ($n = 9$, geometric mean = 8.99 ± 5.19 μg/L) is higher than that in the other two villages ($n = 8$, geometric mean = 6.96 ± 4.28 μg/L); no significant difference was found. The highest Se content was observed in five stream water samples (36.82, 35.39, 33.43, 33.19, 30.6 μg/L) with Se content exceeding 30 μg/L. Thus, Se content in all surface and potable waters were lower than the corresponding standards set by WHO (1986), 40 μg/L and EPA (2002), 50 μg/L.

Probable daily Se intake

Based on the intake rate of each kind of food from the Statistical Yearbook of Shaanxi Province (Shaanxi Bureau of Statistics 2015) and food Se content, Se PDI of Shuang'an habitants was 1067 μg/day (Table 2). PDI of NR habitants
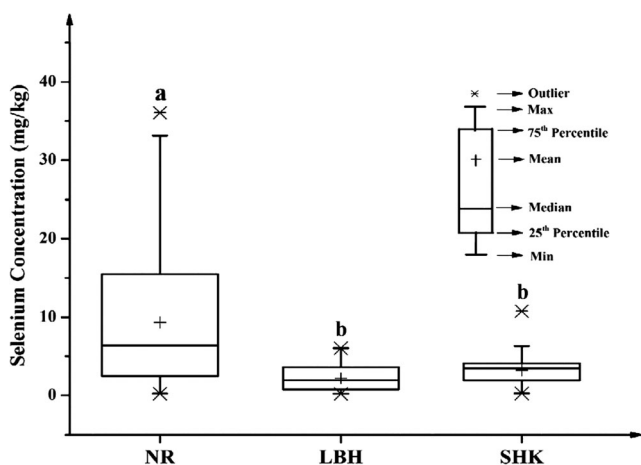
Fig. 2 Se distribution in three villages in Shuang'an of Ziyang County

(1801 μg/day) was much higher than that in LBH (666 μg/day) and SHK (686 μg/day). For local dwellers, major pathways for daily Se intake include cereals, vegetables, and meats. Fruit (4 μg/day) and drinking water (24 μg/day) only slightly contributed to Se PDI.

Se content in hair and blood

The age of randomly recruited participants ($n = 66$) ranged from 16 to 75 years old with an average of 45 years old. Se levels in hair of local habitants varied significantly from 0.59 to 63.05 mg/kg, with a geometric mean of 12.72 ± 13.81 mg/kg. Moreover, hair Se levels of males ($n = 32$, geometric mean = 4.73 ± 3.77 mg/kg) were significantly lower than females ($n = 34$, geometric mean = 14.83 ± 16.76 mg/kg). According to Tan's (1989) classification, 21.21% of the hair samples ($n = 14$) were potentially toxic (0.5–3.0 mg/kg), while 78.79% of the hair samples ($n = 52$) belonged to Se-toxic (>3 mg/kg). Based on the average PDI (1067 μg/day) of local habitants, calculated values of Se content in blood were 1891 and 1649 μg/L through Eqs. (4) and (5), respectively. For habitants living in NR, SHK,

and LBH, the Se content in blood was 2783–2824, 1029–1317, and 1060–1347 μg/L, respectively.

Discussion

Se in soil, plant, and water

Soil Se content of Shuang'an was 3.02 ± 5.16 mg/kg, which was much higher than average Se levels of China (0.173 mg/kg, Tan et al. 2002) and worldwide (0.40 mg/kg, Fordyce 2013), but lower than that from Enshi of China (9.36 ± 18.6 mg/kg) and selenosis-affected areas in India (Jainpur) (6.5 ± 0.3 mg/kg; Jaiswal et al. 2012). Soil Se content of NR in this study was far lower than that reported by Luo et al. (2003, 26 mg/kg) and Fang and Wu (2004, 16.9 mg/kg) in the same place, but it is still significantly higher than that of LBH and SHK. Se in soil mostly comes from weathering of parent material (Qin et al. 2013; Wang and Gao 2001; Yu et al. 2014). In the study area, Se was controlled by the bedrock, a series of pyritic carbonaceous slates, and volcanic substances that created a wide and flat area where the Naore village is located. Because of the thin soil layer, rocks crop out from the floor of the valley and weather into Se-rich soils (Luo et al. 2003).

According to Mei's (1985) study, high Se content in soil accompanies with high Se levels in plants. In this study, 40% of vegetable and 32% of cereal significantly exceeded the Se toxicity standard (1 mg/kg; Tan 1989). In addition, Se content of plant from NR was significantly higher than that from LBH and SHK. Se content in corn (0.81 ± 3.12 mg/kg) was much higher than that from Hubei Enshi (0.43 mg/kg, Huang et al. 2013), Shaanxi Yao County (0.08 mg/kg, Fang et al. 2003), and Heilongjiang Tailai (0.03 mg/kg, Tan and Huang 1991), but lower than that from Hubei Enshi (3.76 mg/kg, Qin et al. 2013) and Venezuela (14 mg/kg, Fordyce 2013). Rice Se levels from this study area were lower than that from

Table 1 Se concentration of plants in Shuang'an area of Ziyang

Food		Number (n)	Max	Min	Geometric mean	Accumulation coefficient
Cereal	Corn	132	17	0.02	0.81 ± 3.12	0.64
	Rice	11	1.63	0.36	0.71 ± 0.39	0.20
	Wheat	1	–	–	0.44	–
Oil plants	Rape	7	0.74	0.04	0.27 ± 0.27	–
Allium	Scallion	2	0.76	0.06	0.41 ± 0.49	–
	Garlic	2	4.02	0.08	2.05 ± 2.79	–
Solanaceous fruits	Pepper	27	7.85	0.04	1.57 ± 1.75	2.83
	Eggplant	5	8.96	0.05	3.99 ± 4.54	4.54
Beans	–	7	1.72	0.03	0.14 ± 0.19	–
Rood tuber	Potato	59	4.67	0.02	0.37 ± 0.84	–
Aquatic vegetables	Lotus	5	0.20	0.11	0.16 ± 0.04	0.12

Table 2 Food consumption and daily dietary selenium intake in NR, LBH, and SHK

	Se standard (µg/kg)[a]	Capita consumption (kg/day or L/day)[b]	Se content (µg/kg or µg/L)[d]			PDI (µg/day)			Se content[d] (µg/kg or µg/L)	PDI (µg/d)
			NR	LBH	SHK	NR	LBH	SHK		
Cereal (DW)	300	0.396	1816	622	705	719	246	279	1257	498
Vegetable (FW)	100	0.282	1590	355	308	448	100	87	761	215
Meat	500	0.064	20,000	6000	6000	500	192	192	7000	224
Egg	500	0.011	9730[c]	9730[c]	9730[c]	107	107	107	9730[c]	107
Potable water	10	3	8.99	6.96	6.96	27	21	21	7.85	24
Total	–	–	–	–	–	1801	666	686	–	1067

DW dry weight, *FW* fresh weight

[a] Se standard from Health Standards of China (GB2762-2005)

[b] Data from Shaanxi Statistical Yearbook (2015)

[c] Data from the research conducted by Hou et al. (1993)

[d] Geometric mean

Venezuela (18 mg/kg, Fordyce 2013) and Hubei Enshi (2.11 mg/kg, Qin et al. 2013), but higher than that from the other areas in China (Huang et al. 2010; Gao et al. 2011) and USA (0.136 mg/kg, Fordyce 2013). Thus, Se levels in plants were high within the scope of China and the world (Table 3).

In addition to the soil Se content, differences in the capacity of Se uptake for plant are another decisive factor which can be reflected by the accumulation coefficient (Table 1). Eggplant and pepper are two kinds of vegetables with accumulation coefficient greater than 1. Corn accumulates more Se than rice; this is due to the different Se uptake ability of plants and the Se speciation of soil. Under the flooded conditions of rice field, selenate is more likely to be reduced to selenite which is less available than selenate (Li et al. 2007). In addition, another environmental influence such as Se level in irrigation water is also the crucial factor that determined Se content in plants.

Se in water is also highly determined by soil Se content. Stream water had an average Se level of 12.35 ± 9.45 µg/L (1.86–36.82 µg/L, Fig. 3), which was comparable to that from the Šobov Slovakia seleniferous area (0.12–32.2 µg/L, Bujdoš et al. 2005) and Punjab, India (0.12–69.5 µg/L, Dhillon and Dhillon 2003), but lower than that in Enshi, China (0.46–275 µg/L, Fordyce 2013). The five water samples with Se content bigger than 30 µg/L were all collected from confluence reaches of streams, where rock erosion is more significant than in the other parts, and more Se erodes into water. However, Se content in potable water was relatively low, and no significant difference between NR village and other two villages was found. The geometric mean of Se content in potable water was 7.85 ± 6.04 µg/L, which is lower than the maximum permissible level (MPL) set by US EPA, WHO, European Union, and Chinese Minister of Health (Table 4).

Table 3 Comparison of food Se content with other selenosis regions in the world

Region	Se content (mg/kg)	Reference
Corn		
Shaanxi, Ziyang	0.81	This study
Hubei, Enshi	0.43	Huang et al. (2013)
Enshi, Hubei	3.76	Qin et al. (2013)
Venezuela	14	Fordyce (2013)
India	13	Sharma et al. (2009)
Rice		
Shaanxi, Ziyang	0.71	This study
Anhui, Shitai	0.70	Bañuelos et al. (2015)
Hubei, Enshi	0.96	Huang et al. (2013)
Hubei, Enshi	2.11	Qin et al. (2013)
Venezuela	18	Fordyce (2013)
Nigeria	2.01	Gbadebo et al. (2010)
Egg plant		
Shaanxi, Ziyang	3.99	This study
Hubei, Enshi	1.04	Huang et al. (2013)
Pepper		
Shaanxi, Ziyang	1.57	This study
Thailand, Bangkok	0.92	Sirichakwal et al. (2005)
India	16.53	Dhillon and Dhillon (1991)
Potato		
Shaanxi, Ziyang	0.37	This study
Hubei, Enshi	0.82	Qin et al. (2013)
Hubei, Enshi	0.28	Huang et al. (2013)
India	1.24	Dhillon and Dhillon (1991)

Fig. 3 Se concentration of water in Shuang'an of Ziyang County
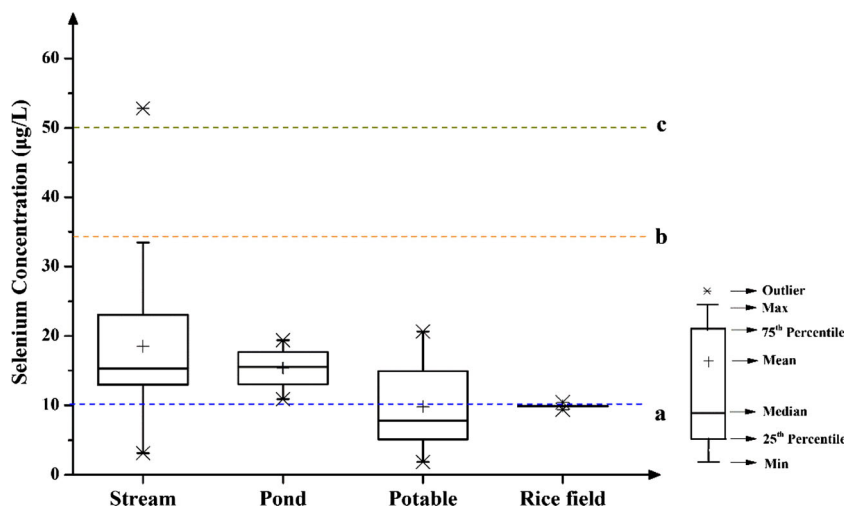
Daily Se intake

The daily Se intake of local residents in NR, LBH, and SHK was 1801, 666, and 686 μg/day, respectively. The average Se intake of studied area was 1067 μg/day, which exceeded tolerable upper Se intake limit (400 μg/day) suggested by WHO (1986) and Chinese Nutrition Society (2001). The average Se intake of this study was higher than those in other Se-rich areas such as Venezuela (200–350 μg/day, Combs and Combs 1986), USA (106 μg/day, Food and Nutrition Board and Institute of Medicine 2000), Greenland (235 μg/day, Hansen et al. 2004), Japan (127.5 μg/day, Miyazaki et al. 2002), India (475–632 μg/day, Hira et al. 2004), Canada (98–224 μg/day, Gissel-Nielsen 1998), and New Zealand (19–80 μg/day, Combs 2001). In contrast, Se intake for residents living in Enshi of Hubei (4990 mg/kg, Yang et al. 1983) was 5 times and 2.8 times higher than the average PDI of this study area and NR village, respectively.

The Se intake is determined not only by the Se content in food but also the diet structure. The staple food of residents living in Ziyang Shuang'an is cereal, and the diet structure is quite unitary. Although corn had lower Se levels than vegetable and meat, 46.65% of inhabitants' daily Se intake comes from cereal consumption. Because of high Se content in meat, it also makes a considerable contribution (20.99%) to daily Se intake. The higher Se in meat may attribute to animals that require more Se than

plants (Klapec et al. 2004). The contribution of eggs and potable water to daily Se intake is less than 15% (Fig. 4). These findings were consistent with those from a previous research where cereals were the major contributors of Se intake, followed by fish and meat, with vegetables and fruits contributing the least (Choi et al. 2009; Gao et al. 2011). However, research on Se PDI in Saudi Arabia showed that daily Se intake was mainly from meat and that eggs and cereals only account for approximately 16% of daily Se intake, and these differences may be due to variable dietary habits and distinct Se levels in consumed food (Fordyce 2013; Yu et al. 2015).

Risk assessment of Se poisoning

The average HQ of this area was as high as 4.34, which was calculated using the average daily selenium intake and recommend daily allowance. HQ for NR, LBH, and SHK was 7.50, 2.78, and 2.86, respectively; selenosis area of NR was nearly three times of the other two villages. All these HQ values were much higher than 1 and the results in Indian Se-rich areas (0.98, Dhillon and Dhillon 2015). The high HQ value indicated that residents living in this area were at a huge risk of Se toxicity, especially for habitants living in NR village. Se in hair, which acts as the terminal of the rock–soil–plant–human body ecosystem, can verify the Se status of people (Gao et al. 2011; Chen et al. 2013). The geometric mean of hair Se level

Table 4 Standards for drinking water

Source	Se level	Reference
This study	7.85 ± 6.04 μg/L	–
WHO (Guidelines for Drinking-Water Quality 4th Ed)	40 μg/L	WHO (2011)
EPA	50 μg/L	EPA (2012)
Regulation for Europe and French (Directive 98/93/CE-03.11.98)	10 μg/L	Caussat et al. (2007)
Sanitary Standard for Potable Water Quality of China	10 μg/L	GB5749-2006

Fig. 4 Food contribution to daily Se intake for residents in Shuang'an of Ziyang County
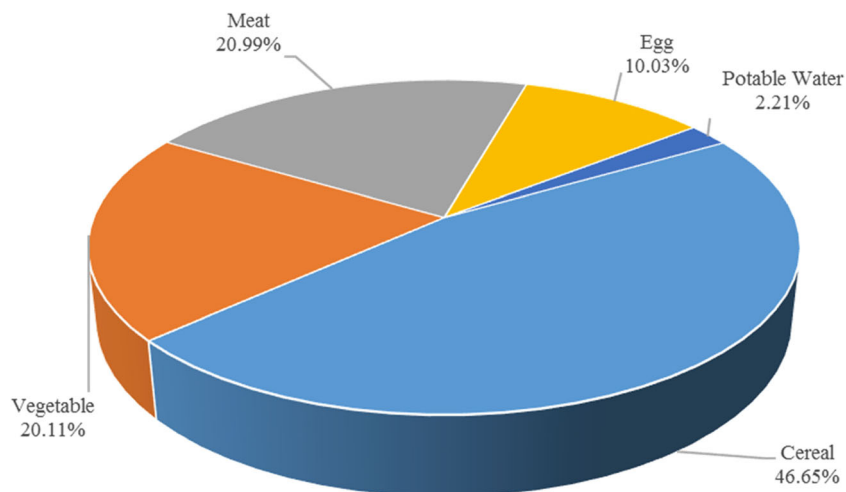
in local inhabitants was 7.05 ± 13.81 mg/kg. The result was lower than that in Enshi Se-rich areas (21.5 ± 20 mg/kg, Xia et al. 2000) and India seleniferous areas (28.21 mg/kg, Dhillon and Dhillon 1991). However, Se content in human hair was two times of toxic standard suggesting by Tan (1989), which further indicates Se poisoning risk in study area. In addition, Se content of female hair was threefold of that in male hair, which is in agreement with previous studies (Hira et al. 2004), indicating that high Se exposure levels in women would be a matter of concern in future Se risk assessments.

In addition to hair Se, blood Se is also a favored biomarker when comparing the Se status between different living conditions (Pestitschek et al. 2013). According to WHO (1986), Se toxicity was observed in hair and nails at whole blood Se levels ranging from 1020 to 1854 μg/L. EPA sets the NOAEL of Se content in blood at 1000 μg/L and the lowest observable adverse effect level (LOAEL) at 1350 μg/L for blood Se (EPA 1992). Calculation of blood Se content through daily Se intake and hair Se content showed that the blood Se status of inhabitants from NR (2783–2824 μg/L) was twice as high as that in LBH (1029–1317 μg/L) and SHK (1060–1347 μg/L), indicating that habitants living in NR were facing a greater risk of Se poisoning. The calculated blood Se status of local residents was much higher than that of NOAEL and results in other Se-rich areas such as Amazon (284.3 μg/L, Lemire et al. 2006; 228.4 μg/L, Lemire et al. 2012) and Greenland (488 μg/L, Hansen et al. 2004), which further confirmed the huge Se poisoning risk for local inhabitants.

Despite the non-negligible risk reflected by daily Se intake and Se content in hair and blood, there have been no cases of Se poisoning in the last 30 years, which was in agreement with the result in Greenland and Brazilian Amazon populations despite their high Se intake (Hansen et al. 2004; Lemire et al. 2012). Previous research found that human exposed to high environmental Se levels may tolerate higher Se contents through homeostatic adjustment (ATSDR 2003; WHO 1986) and may even develop

mechanisms to take advantage of such the high Se intake (Mattson 2008). Zhu et al. (2009) also suggested that because SeMet is the major Se species in plant Se, which is less toxic than SeCys and inorganic Se, Se poisoning may have disappeared. The Institute of Medicine (IOM 2000) emphasized that SeMet may not entirely undergo homeostatic regulatory processes as a consequence of non-specific accumulation. As such, it may act as a storage pool for Se and play an important role in adjusting Se status balance. Although the risk of Se toxicity may have been reduced, further research should be performed to examine Se species, their conversion in staple food, and Se intake of humans.

Conclusion

High Se levels were observed in the soil samples from Ziyang Shuang'an and distributed unevenly; approximately 72% of the soil was classified as Se toxic. The Se content in plants varied and exhibited an ability to accumulate, with a geometric mean of 0.76 ± 2.51 mg/kg. Se content of soil and plant in NR was significant higher than that in adjacent villages LBH and SHK. Stream water yielded the highest Se level, with a geometric mean of 12.35 ± 10.46 μg/L. The Se content in potable water (7.85 ± 6.04 μg/L) was lower than the standard of WHO and EPA. The high Se content in hair and the calculated daily Se intake indicated that the inhabitants of Shuang'an in Ziyang were at a high risk of Se poisoning, especially for habitants of NR.

Selenosis was not evident in our study population, whose Se intake is obtained from traditional diet. Nevertheless, local government should consider agronomic measures to regulate the availability of Se. Increased awareness and enhanced knowledge are keys to modifying the single-diet structure, reducing dependence on cereals, and increasing the amount of imported food.

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